

Antenna-coupled Photon Emission from hexagonal Boron Nitride Tunnel Junctions

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The ultrafast conversion of electrical to optical signals at the nanoscale is of fundamental interest for data processing, telecommunication and optical interconnects. However, the modulation bandwidths of semiconductor LEDs are limited by the spontaneous recombination rate of electron-hole pairs and the footprint of electrically-driven ultrafast lasers is too large for practical on-chip integration. A metal-insulator-metal (MIM) tunnel junction approaches the ultimate size limit of electronic devices and its operating speed is fundamentally limited only by the tunneling time. Here we study the conversion of electron energy – localized in vertical gold–h-BN–gold tunnel junctions – into free space photons, mediated by resonant slot antennas. Optical antennas efficiently bridge the size-mismatch between nanoscale volumes and far-field radiation and strongly enhance the electron-photon conversion efficiency. We achieve polarized, directional and resonantly enhanced light emission from inelastic electron tunneling and establish a novel platform for studying the interaction of electrons with strong electromagnetic fields. Our results pave the way for the further development of nanoscopic sources of light enabled by the combination of nanophotonic design principles and nanoelectronics.

Optical nanoantennas are elements that couple free space radiation to material structures with volumes that are orders of magnitude smaller than the wavelength of a visible photon (400 to 700 nm) [1–5]. Incoming radiation is converted into localized surface plasmon polaritons (LSPs) – time-harmonic oscillations of the free electron gas – that concentrate electromagnetic energy into ultrasmall volumes [6]. Many optical antennas owe their design to their macroscopic radiofrequency predecessors like linear dipole antennas [2, 7], Yagi-Uda antennas [8] or bow-tie antennas [9].

Classical antennas are used to generate radiofrequency (RF) waves by driving them at the respective frequency electrically or – in reverse – to generate RF electrical signals from incoming electromagnetic radiation. Optical antennas on the other hand have been mostly operated on a “light-in / light-out” basis [4]. This paradigm has recently started to shift towards the integration of optical antennas in optoelectronic devices for photovoltaics [10], photon detection [11, 12] and surface plasmon excitation [13]. Here we report on the realization of solid-state, light-emitting tunnel devices based on arrays of electrically driven optical antennas.

Applying a voltage to a metal-insulator-metal junction yields a current due to electrons tunneling through the insulator from occupied states in one electrode to unoccupied states in the other electrode. Generally speaking, the majority of electrons tunnel elastically, maintaining their energy during the tunneling process. The excess energy of the “hot electron” is subsequently thermalized. In 1976, Lambe and McCarthy found that surface plasmon modes in metal-insulator-metal (MIM) tunnel junctions increase the likelihood of inelastic tunneling, a process in which a tunneling electron

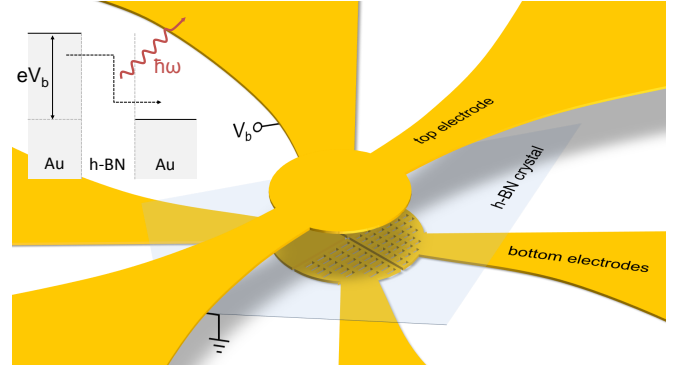


FIG. 1. Expanded illustration of the sample configuration. Devices consist of a vertical stack of segmented, nanostructured gold bottom electrodes, few-layer hexagonal boron nitride (h-BN) and a common gold top electrode. We observe and analyze light emission due to inelastic electron tunneling (see inset), controlled by the optical, geometrically defined, properties of the devices. The inset shows the excitation of a photon with energy $\hbar\omega$ by inelastic electron tunneling.

excites a surface plasmon, cf. inset of Fig. 1, which may subsequently decay into detectable far-field radiation [14]. This effect has since been studied experimentally in macroscopic solid-state tunnel junctions [15, 16], and the scanning tunneling microscope (STM) [17–21], as well as theoretically [22–25]. One major appeal of investigating inelastic electron tunneling is its potential speed. Since it does not rely on intermediate excitations such as electron-hole-pairs, the operating speed of devices is fundamentally only limited by the tunneling time of electrons through the junction, which is of the order of tens of femtoseconds [26], hence opening up applications in ultrafast optoelectronics and communication.

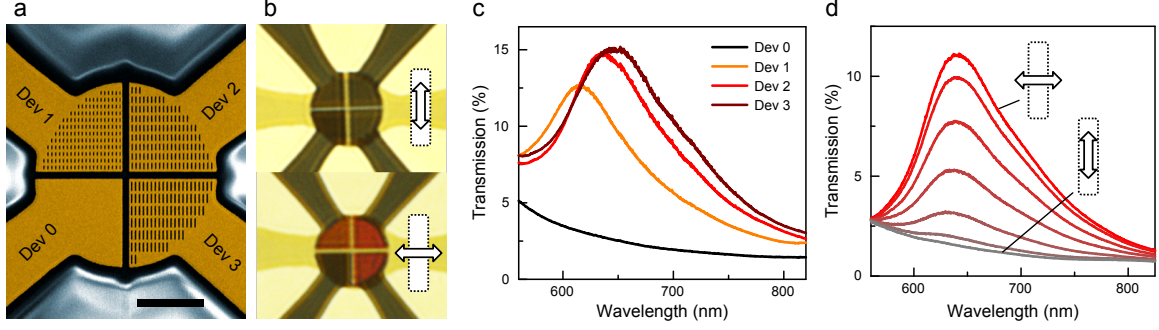


FIG. 2. Structural and optical properties of the tunnel devices. (a) False color SEM image of a nanostructured bottom electrode. Sizes of the slots are $150 \times 50 \text{ nm}^2$, $250 \times 50 \text{ nm}^2$ and $350 \times 50 \text{ nm}^2$ for Devices 1, 2 and 3, respectively. Scale bar $2.5 \mu\text{m}$. (b) Polarized optical transmission micrographs of the final device. Light polarized parallel to the short axis of the slots generates distinct color variations amongst the different electrodes. The colors disappear for light polarized along the long axis. (c) Transmission spectra of the four devices, quantifying the qualitative observation from (b). The antenna arrays give rise to characteristic spectral resonances. (d) Transmission spectra of Dev 3 as a function of polarizer angle from 0° (parallel to the short axis) to 90° (parallel to the long axis) in steps of 15° .

The tunnel barrier of top-down fabricated devices is commonly based on the oxides grown on one of the electrodes (e.g. aluminum oxide) or on insulating films deposited by atomic-layer deposition (ALD) or sputtering. Unfortunately, due to grain formation and defects, these tunnel barriers are unstable and operation over extended time at ambient conditions is not possible. Here, we investigate light generation via inelastic electron tunneling in microscopic tunnel devices comprised of gold (Au) and hexagonal boron nitride (h-BN). h-BN provides a high-quality tunnel barrier due to its crystallinity [27, 28] and a large bandgap of $\sim 6 \text{ eV}$ [29]. We find that light emission can be enhanced and spectrally controlled by nanostructuring one of the electrodes. Light emission and optical properties of the device are found to be closely related.

Fig. 1 illustrates the sample configuration. We photolithographically define a circular gold bottom electrode ($1 \text{ nm Ti} / 50 \text{ nm Au}$) that is subsequently nanostructured by focused ion beam milling (FIB). The bottom electrode is segmented into four quarter-circle electrodes, which are wired individually as shown in Fig. 2a. One electrode remains unstructured (Device 0) to serve as a reference, while the remaining three electrodes are structured into arrays of optical antennas in the form of rectangular slots. The distance between slots is kept constant at $\sim 100 \text{ nm}$ along both the long and short side of the rectangles while the nominal size of the slots is $150 \times 50 \text{ nm}^2$, $250 \times 50 \text{ nm}^2$ and $350 \times 50 \text{ nm}^2$ for Devices 1, 2 and 3, respectively. Exfoliated few-layer h-BN (here: 9 atomic layers, or $\sim 3 \text{ nm}$) is transferred on top of the bottom electrode. The device is finalized by the fabrication of a common gold top electrode (15 nm Au) via photolithography. Details on the fabrication process can be found in Methods and the Supplementary Information.

Fig. 2b-d depict and analyze the optical properties of the final devices. The transmission of light through the devices strongly depends on its (linear) polarization, as shown in Fig. 2b. Observing light polarized along the short axis of the slots results in a strong color contrast between devices. This contrast vanishes when observing light polarized along the long axis. Since Device 0 is unstructured its transmission does not depend on polarization. The colors of the electrodes suggest that the slots enhance transmission in a certain spectral range. This observation is quantified in the transmission spectra $T(\lambda)$ shown in Fig. 2c. The transmission spectrum of Device 0 falls off continuously towards longer wavelengths, in agreement with analytical calculations (see Supplementary Information). All structured devices exhibit characteristic resonances and enhanced transmission at wavelengths corresponding to the observed colors in Fig. 2b. The transmission peak shifts towards longer wavelengths as the aspect ratio of the slots is increased in accordance with Ref. [30, 31]. Fig. 2d exemplarily shows the change in the transmitted spectrum as a function of polarizer angle. The resonance completely disappears when the analyzer is oriented along the long axis of the slots. The transmission follows a $\cos^2 \theta$ dependence (see Supplementary Information) [32].

Next, we discuss the properties of photon emission induced by inelastic electron tunneling. The top-most image in Fig. 3a shows a real-space map of the light emitted from the unstructured reference device, recorded with a constant tunnel bias voltage of $V_b = 2.5 \text{ V}$. The majority of the emitted light originates from the edges of the device while the remainder of the device area appears dark. The spectrum of the emitted light, shown in Fig. 3b, is broad and without distinct features, falling

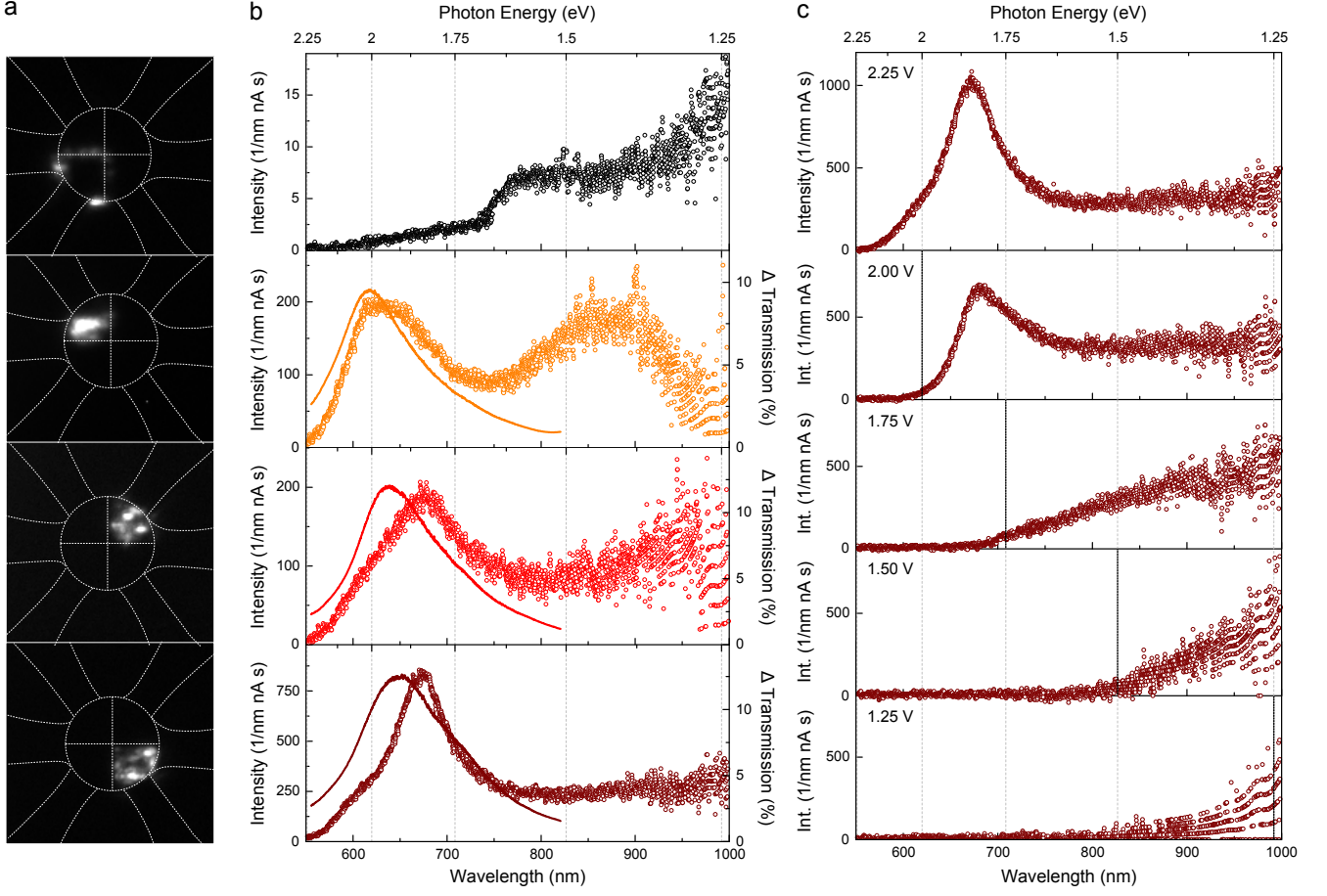


FIG. 3. Light emission from antenna-coupled tunneling devices. (a) EMCCD images of the light emitted from the four devices at a constant applied bias of $V_b = 2.5\text{V}$. (b) Spectral intensity of the emitted light for all four devices at $V_b = 2.5\text{V}$ (open circles) in comparison with the optical transmission enhancement, i.e. $\Delta T_i(\lambda) = T_i(\lambda) - T_0(\lambda)$ (solid lines). Resonance peaks are observed in all structured devices. The peaks shift towards longer wavelengths with increasing aspect ratio of the slots. (c) Light emission spectra for Dev 3 as a function of the applied bias. The dark dashed lines indicate the respective cut-off photon energy given by $\hbar\omega = |eV_b|$.

off continuously towards shorter wavelengths. In the absence of slots, as is the case for the reference device, inelastically tunneling electrons predominantly interact with surface plasmon polaritons (SPPs) associated with the metal-insulator-metal (MIM) configuration [33, 34]. The dispersion relation of these modes can be calculated analytically (see Supplementary Information). The field of MIM-SPPs is strongly confined between the two metal layers, giving rise to field momenta $\hbar k$ that are much larger than the photon momentum $\hbar k_0$ in free space ($k_0 = 2\pi/\lambda$, with λ being the photon wavelength). We find $\hbar k \sim 10\hbar k_0$ for $\lambda = 1000\text{nm}$ increasing to $\hbar k \sim 30\hbar k_0$ for $\lambda = 500\text{nm}$. The propagation lengths of these modes vary between $\sim 350\text{nm}$ for $\lambda = 1000\text{nm}$ and $< 10\text{nm}$ for $\lambda = 500\text{nm}$. In the absence of discontinuities, these modes cannot couple to free-space radiation because of the large momentum mismatch. However, scattering due to surface roughness or edges provides the momentum required to overcome this mismatch. Hence

we only observe light emission from the edges of Device 0.

In contrast, all devices with nanostructured bottom electrodes show light emission from the entire device area (Fig. 3a, bottom three panels). As seen in the corresponding spectra in Fig. 3b, the antenna-coupled devices exhibit strongly enhanced light emission intensity, especially in the spectral domain of the transmission peaks discussed previously. Compared to the unstructured device (Device 0) the electron-to-photon conversion efficiency is enhanced by nearly two orders of magnitude. A direct comparison of the transmission and emission spectra shows strong similarities in spectral width and shape. However, the peaks observed in the light emission appear to be slightly red-shifted compared to the peaks in transmission. Numerical simulations, which will be discussed later on, suggest that this spectral shift is caused by the spectral dependence of electron-radiation coupling mediated by the slot antennas. The spatial non-uniformity of the emitted light intensity is due

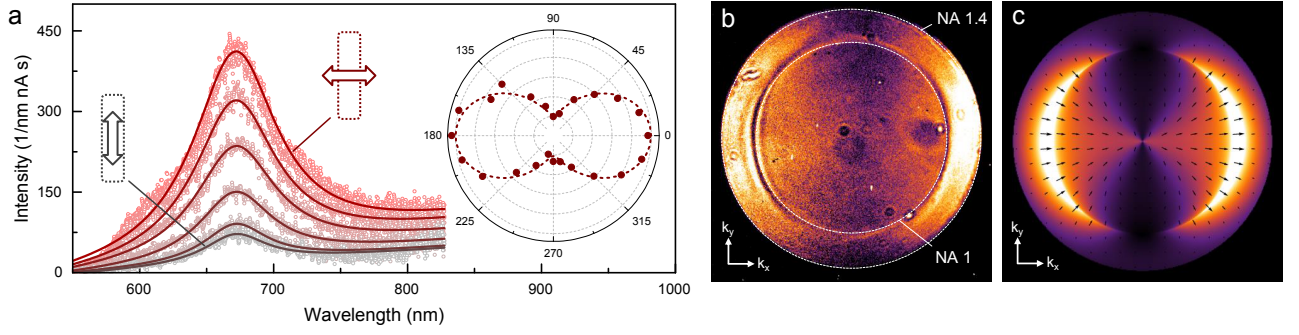


FIG. 4. Dipolar characteristics of the emitted light. (a) Light emission spectra of Dev 3 at $V_b = 2.5\text{V}$ as a function of the analyzing polarizer angle θ from 0° (parallel to the short axis) to 90° (parallel to the long axis) in steps of 15° . The superimposed curves are Lorentzian fits on a linear background. The inset shows a polar plot of the spectrally integrated light emission intensity fitted by a $\cos^2 \theta$ function. (b) Photon momentum distribution in the backfocal plane of the objective for Device 3 at $V_b = 2.5\text{V}$. (c) Analytically calculated photon momentum distribution for a magnetic dipole m_y in air oriented parallel to and at a distance of 25 nm from a glass ($n=1.52$) interface (objective NA 1.4). Vectors indicate direction and magnitude of the real part of the electric field at the respective vector positions.

to fabrication-related contamination at the Au-h-BN interfaces and roughness of the polycrystalline electrodes to which the atomically flat h-BN crystal cannot conform perfectly [28]. Photon emission can be made spatially more uniform by using monocrystalline gold electrodes [35].

As reported by Lambe and McCarthy, inelastic electron tunneling results in broad-band light emission with a high-frequency cutoff given by

$$\hbar\omega_{\max} = |eV_b|, \quad (1)$$

with $\hbar\omega_{\max}$ being the photon energy, e the electron charge and V_b the tunnel bias voltage [14]. According to equation (1), the antenna-specific resonances should only be excitable if the applied bias exceeds the energy of the antenna mode. As shown in Fig. 3c, this behaviour is indeed observed. The figure renders light emission spectra for Device 3 as a function of applied bias V_b . The experimental data shows good agreement with equation (1).

To understand the role of the slot antennas in the light generation process, we studied the polarization and radiation characteristics of the emitted light. Fig. 4a shows emission spectra for Device 3 as a function of polarizer angle. The light intensity is maximized when the polarizer is oriented parallel to the short axis of the slots. The inset of Fig. 4a shows a polar plot of the integrated intensity as a function of polarizer angle θ fitted by a $\cos^2 \theta$ function. For a polarizer angle parallel to the long axis of the slots we observe a $> 80\%$ extinction. Hence we find that light emitted from the electrically driven tunnel devices behaves analogously to the light transmission through the device. Fig. 4b shows the photon momentum distribution in the back focal plane of the microscope objective (NA 1.4). Light is preferentially emitted in x -direction, i.e. parallel to the

short axis of the slots. The measured polarization and the back focal plane intensity distribution qualitatively agree with the radiation generated by a magnetic dipole over a dielectric substrate [36] and oriented along the long axis of the slots (c.f. Fig. 4c). These results are in agreement with the notion that slots can be viewed – in terms of Babinet’s principle – as the counterpart of linear rod antennas, radiation from which is dominated by an electric dipole mode along the long axis of the rod [37]. However, as will be discussed later, slots also support electric dipole resonances parallel to their short axis. The dark ring at an angle slightly larger than NA 1 observed experimentally is not reproduced by analytical calculations. This is most likely caused by coupling to surface plasmon polaritons at the gold-air interface, which are non-radiative due to a total metal thickness of nominally 65 nm separating this interface and the lower half-space, where radiation is detected.

To establish a quantitative understanding of the electron-to-photon conversion efficiency and its spectral dependence, we express the spectrum of the emitted power as

$$p_{em}(\omega) = \hbar\omega \Gamma_{e-p}(\omega) G_{antenna}(\omega) \quad (2)$$

with Γ_{e-p} being the electron-to-plasmon conversion rate and $G_{antenna}$ the antenna gain. Γ_{e-p} is described by Fermi’s golden rule [19, 24, 38] and depends on the electronic density of states in the electrodes as well as the number of plasmonic modes that the electrons can couple to [25]. The latter corresponds to the local density of electromagnetic states (LDOS) in the MIM tunnel gap. The antenna gain $G_{antenna}$ denotes the efficiency of the slot antennas at converting the energy of MIM plasmons into free space radiation. We next discuss the spectral dependence of Γ_{e-p} and $G_{antenna}$ for a $150 \times 50 \times 50 \text{ nm}^3$ slot antenna, additional data

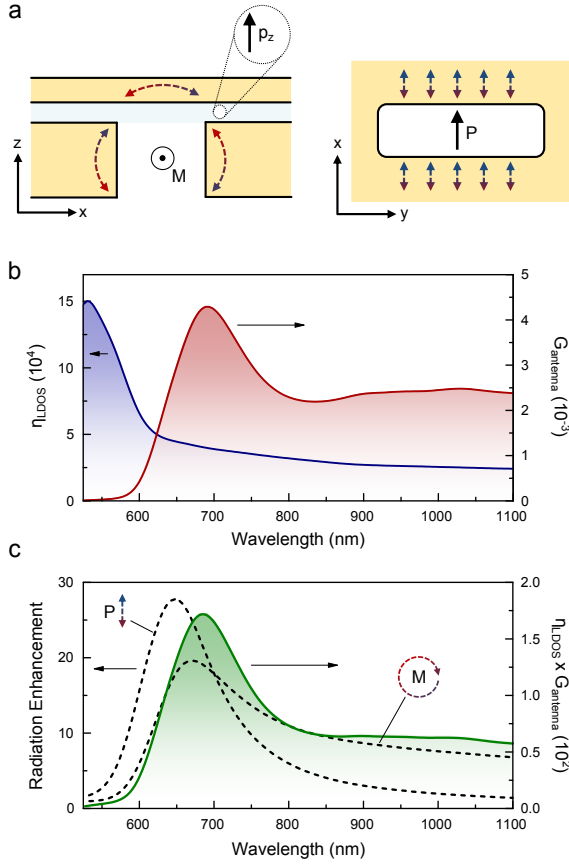


FIG. 5. Finite-element calculations of the emission spectrum. (a) Graphical illustration of dipole placement (p_z) and current distributions (dashed arrows) associated with magnetic (M, left) and electric (P, right) dipole resonances. (b) Local density of electromagnetic states (LDOS) and corresponding antenna gain spectrum. The LDOS is normalized by the LDOS in a homogeneous dielectric environment. (c) Total radiation enhancement ($\eta_{\text{LDOS}} \times G_{\text{antenna}}$) in comparison with electric and magnetic dipole resonances of a slot antenna. The size of the slot is $150 \times 50 \times 50 \text{ nm}^3$.

can be found in the Supplementary Information.

We derive the LDOS seen by the tunneling electrons by numerically calculating the total dissipated power (P_{tot}) of a point dipole p_z placed in the tunnel junction. The normalized LDOS (η_{LDOS}) is then obtained by [36]

$$\eta_{\text{LDOS}} = \frac{P_{\text{tot}}}{P_0}, \quad (3)$$

where P_0 is the radiated power of a dipole of equal dipole moment in a homogeneous dielectric environment. Fig. 5b shows the results for a dipole placed at a distance of 5 nm from the edge of an antenna slot (c.f. Fig. 5a). By varying the lateral position of p_z in the tunnel gap we can map out η_{LDOS} as a function of proximity to the antenna slot (see Supplementary Information). A large fraction of the LDOS is associated with MIM-SPP modes, which are ultimately dissipated to heat in the

absence of antenna coupling. The conversion of SPP modes to free-propagating radiation is most efficient for dipoles that are close to the antenna slot.

In terms of the radiated power P_r , the antenna gain can be expressed as

$$G_{\text{antenna}} = \frac{P_r}{P_{\text{tot}}}. \quad (4)$$

Fig. 5b renders G_{antenna} as a function of wavelength for the dipole near the edge of an antenna slot. We find that the antenna converts SPP modes to propagating radiation most efficiently in a narrow spectral range centered at $\lambda = 690 \text{ nm}$. This antenna resonance can be associated with radiative magnetic and electric dipole modes, as discussed below. The total radiation enhancement is calculated as $\eta_{\text{LDOS}} \times G_{\text{antenna}} = P_r/P_0$ and is shown in Fig. 5c. The spectral shape agrees well with our measurements shown previously in Fig. 3b.

Electromagnetic radiation from an aperture can be described by a multipole series, with the leading terms for small apertures being magnetic and electric dipoles. As illustrated in Fig. 5a, we find that a slot antenna mainly enhances radiation from an electric dipole oriented along the short axis (P), and from a magnetic dipole oriented parallel to the long axis (M) of the slot, in agreement with recent cathodoluminescence measurements [39]. Furthermore, we find that the resonances of both magnetic and electric emission modes red-shift with increasing slot aspect ratio, in accordance with both experimental transmission and light emission results (see Supplementary Information). The dashed curves in Fig. 5c show calculated enhancement spectra for P and M. The radiation of both dipoles is resonantly enhanced, with the M resonance being slightly red-shifted with respect to the P resonance. Furthermore, the M resonance is asymmetric with a long-wavelength tail due to strong coupling to MIM-SPPs, the damping of which increases with decreasing wavelength.

Returning back to equation (2), we now realize that the spectral dependence of the antenna-coupled tunneling device is dictated by the intricate interaction of tunneling electrons and electromagnetic modes supported by the slot antennas. As discussed earlier, our experimental results indicate that the enhanced light emission is mediated primarily by the magnetic dipolar antenna mode. This experimental observation is substantiated by the spectral overlap of M with the radiative LDOS as well as the symmetry of the mode shown in Fig. 5b. Furthermore, again for reasons of symmetry, linearly polarized far-field radiation primarily couples to in-plane electric dipole modes, i.e. the resonant enhancement of transmission through the device is mediated by the electric dipole mode P. These conclusions are supported by the observed spectral shift

between the transmission and emission peaks discussed earlier, which we also find between electric dipole mode spectrum P and the radiative LDOS in the tunnel gap (c.f. Fig. 5c).

Further studies will be aimed at the increase of efficiencies through alternative antenna designs (e.g. patch antennas [40] or nanocube antennas [41]) and improvements in fabrication and materials. For example, MIM propagation lengths may be increased through the use of monocrystalline metals [35] or alternative materials [42–44]. Higher efficiencies and spectrally narrower resonances can also be achieved through MIM gap resonances (see Supplementary Information). Equivalent circuit models will help to improve impedance matching between the tunnel junction and free-space, thereby improving the device efficiency further [1–3].

In conclusion, we have demonstrated optical antenna mediated conversion of electronic into optical energy in a solid-state system. We find that arrays of slot antennas exhibit geometrically tunable, polarization-sensitive resonances that facilitate enhanced device transmission. While light emission from unstructured devices is weak and largely limited to the edges of the device, slot antenna arrays emit light from the entire device area. The emitted light is strongly polarized along the short axis of the slots in correspondence with the polarization dependence of the transmission. Peaks in transmission and light emission for the same device are spectrally shifted with respect to each other. Numerical simulations suggest that the enhanced transmission of far-field radiation is mediated by the electric dipole component of the slot resonance. On the other hand, it is the magnetic dipole resonance that couples localized plasmons in the tunnel junction to free-space radiation.

We have developed and presented a platform for the study of the interaction of low-energy electrons with localized fields in nanoscale volumes. The vertical design of slot antennas allows for sub-nanometer control of gap-size and configuration, a goal which is difficult to achieve in traditional in-plane antenna designs. We show for the first time deterministic light emission due to inelastic electron tunneling in a solid state system using a two-dimensional atomic crystal, namely hexagonal boron nitride. In particular, the exploration of van der Waals heterostructures [45] with modified electronic properties may lead to significant improvements in efficiencies and enable novel device functionalities. By reciprocity, the geometry presented in this study is also capable of converting far-field radiation into localized energy, which is of interest for photovoltaics, photodetection and optical sensors.

METHODS

Sample Fabrication

Devices are fabricated on commercially available glass coverslips ($22 \times 22 \times 0.13 \text{ mm}^3$). Both bottom and top electrodes were fabricated by standard UV photolithography, electron-beam evaporation of titanium and gold targets, and subsequent lift-off. Bottom electrodes were nanostructured using a dual-beam FEI FIB Helios 600i.

h-BN crystals were grown as described in Ref. [46]. Few-layer h-BN atomic crystals were exfoliated on silicon wafers with a 280 nm oxide layer using the scotch-tape micromechanical cleavage technique [47, 48]. Crystals were identified by means of optical microscopy as well as atomic force microscopy using a Bruker Innova AFM in tapping mode.

The transfer was carried out as follows: The Si/SiO₂ substrate containing the h-BN crystals is partially covered with a commercial PDMS film (Gel-Film® PF-40-X4 supplied by Gel-Pak) in such a way that small margins along the edges of the substrate are left exposed. The stack is subsequently floated on deionized water, the exposed (hydrophilic) Si/SiO₂ surface is wetted such that a meniscus is formed around the hydrophobic PDMS and heated to 90 °C. After ~ 30 min the stack is completely immersed in water and the PDMS film is peeled-off under water. During this process the majority of h-BN crystals gets transferred from the Si/SiO₂ to the PDMS. The PDMS film, carrying the h-BN crystal, is subsequently aligned manually with the bottom electrode using a SUSS MJB4 mask aligner, attached to a quartz carrier, and brought into contact with the bottom electrode. As the PDMS film is released from the sample, the h-BN crystal gets transferred onto the bottom electrode. See Supplementary Information for AFM and optical images of the transfer process.

Samples were annealed in a 400 sccm flow of 95 % Argon and 5 % Hydrogen at 300 °C for ~ 3 h before and after transfer.

Sample Characterization

All measurements were carried out on a customized, inverted Nikon TE300 microscope. For transmission measurements light from a supercontinuum source (NKT SuperK EXTREME) was focused onto the devices through a top objective (NA 0.8). Transmitted light as well as light emitted by the devices was collected using an oil-immersion objective (NA 1.4). Spectra of the transmitted as well as emitted light were acquired with an Acton SpectraPro 300i spectrometer. Transmission spectra are normalized by spectra acquired on glass,

which is assumed to correspond to the theoretical value of 96 %. Units of light emission spectra are not arbitrary. They are normalized by the system efficiency, acquisition time and the average current flowing through the device during acquisition. The system response was calibrated using an OceanOptics HL-2000-CAL calibration lamp. Real space as well as back-focal plane images of the emitted light were acquired using an Andor iXon Ultra EMCCD camera. As a voltage source and for electrical measurements we used a Keithley 2602B source meter.

Numerical Simulations

Numerical finite-element simulations were carried out with COMSOL Multiphysics 4.4. We assumed refractive indices of 1.52 for glass and 1.8 for h-BN [49]. Empirical values were used for the dielectric function of gold [50]. The 1 nm adhesion layer of the lower electrode was not taken into account. Full-field simulations of the electromagnetic fields were performed for electric / magnetic point dipoles at the positions mentioned in the main text. The radiated power was extracted by integrating the time-averaged Poynting vector over the simulation boundaries in the lower and upper half-spaces. The non-radiative energy loss rate is calculated by integrating the total power dissipation density within the metallic domains. Perfectly matched layers were used as simulation boundaries.

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AUTHOR CONTRIBUTIONS

L.N., P.B. and M.P. conceived the research. M.P. fabricated the samples and carried out the numerical simulations. M.P. and P.B. measured the samples. A.J. developed the transfer technique. K.W. and T.T. synthesized the h-BN crystals. M.P, P.B. and L.N. discussed results and co-wrote the paper.